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13. ABSTRACT (*Maximum 200 words*)

This is the final report of the above named project. During the grant period, significant progress has been made on the problem of the stopping (linear energy transfer) of swift, charged particles in matter. In particular, the electronic structure of the ultra thin foils have been determined. Their stopping has been calculated. These pilot calculations have indicated that further work along these lines will produce useful results.

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Theoretical and Numerical Prediction of Stopping Properties of Counterpart Thin Films and Solids

Final Report

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May 8, 1991

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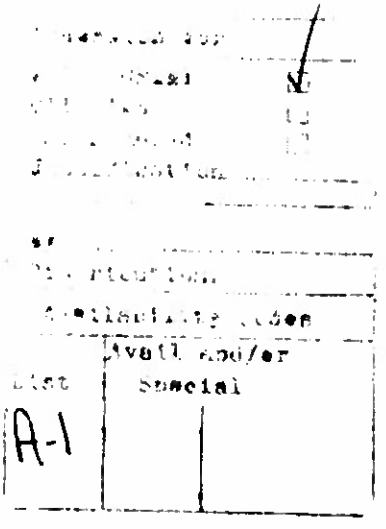

1. Statement of the Problem

The problem this research addressed is the prediction and description of stopping of swift, massive, charged particles in atoms, molecules, ultra-thin material foils and bulk solids. Although the foils and eventually solids were our main concern, it was necessary to consider atoms and molecules along the way, as they introduce progressively more complicated models and experimental situations that can be used to test and calibrate our methods. The methods used here are based on the Kinetic Theory of Stopping (KT), which requires knowledge of the velocity density of scatterers (the target electrons) and the mean excitation energy of the target electrons. We proceed along two general lines, which we refer to as "first principles" and "semi-empirical."

2. Summary of Results

We use the phrase "first principles" here to mean that we do not use any experimental information in the description of the stopping of films, rather we compute everything within a well defined scheme containing no adjustable or fit parameters. [We note that the orbital local plasma approximation (OLPA) used in some of the work is somewhat arbitrary, but does not contain any experimental information and has the proper uniform gas limit. See discussion below.] In addition to being without empirical components, we desire that the development be consistent in its antecedents. In our scheme, this means that the various components of the development are all based, in one way or another, on the electron density of the target system.

In order to do this, we need put together several theoretical and computational pieces, none of which is "off the shelf," and thus each must be developed such that all pieces are consistent. We have attempted to develop several of the pieces in parallel during the contract period. We review the component efforts here.

The central ingredient of the development is the stopping theory itself. We use the Kinetic Theory of Stopping (KT) in its orbitally resolved implementation, which requires a set of mean excitation energies and the target electron velocity density. The first implementation of this theory, for atomic targets, and for the Bethe/Born term only, was made in this laboratory[1], and subsequent applications to atomic targets have given good results when compared to other computations and to the available experimental data. Recently we have been working on a polarization propagator calculation of Generalized Oscillator Strengths (GOS) for atoms and molecules (all that is possible at the moment). Integration of the GOS over momentum transferred gives the stopping cross section directly. Thus a calculation of the stopping of selected atomic and molecular targets *via* the GOS and *via* the KT will give a direct and definitive test of the KT and its orbital decomposition. Such a test has not been made up until now as it has not been possible to calculate the GOS's generally and reliably. We expect this work to be published soon[2].

The KT is compatible with the local density approximation (LDA) to density functional theory (DFT) description of the electronic structure of the film as they are both theories based on the ground state electron density (in real or momentum space) of the target. It is this scheme that we have been developing for calculation of the electronic structure of the target, because of the focus upon ordered thin film targets.

In this context the first challenge is to get a reasonable theoretical description of the electronic structure of the thin film in order that we can determine the target electron velocity density.

With many others, we have argued that the LDA/DFT is a state-of-the-art method to use in this endeavour. During the past contract period, the computational techniques for thin films, which wereconceived in this laboratory in the early 1980's[3], have been developed to the point that they produce reliable, predictive results. That is, we can now calculate the electronic structure and ground state geometry of films of several atomic layers and with complicated unit cells, and have done so for diverse systems of varying complexity.[4] A preliminary report on the stopping of protons in several thin films, calculated using densities calculated in this way, has been accepted for publication, and will soon be out[5] (*vide infra*).

The second challenge to theory and computation is the central material parameter that enters the KT, the mean excitation energy, I, of the target. I is defined as the logarithmically weighted dipole oscillator strength distribution. To calculate I directly from its definition requires a knowledge of the complete dipole oscillator strength distribution of the target, a quantity which cannot at present be calculated from first principles either for thin films or for solids. Consequently we have chosen to use an orbital formulation of the local plasma approximation (LPA) in our formulation. Although not derivable from first principles, the LPA is one of the few accessible methods for calculating mean excitation energies in extended systems, and is consistent with the rest of our formulation in that it is also based on the target electron density (in real space density). Introduction of the orbital version of the LPA (OLPA) leads to a choice of models, as it is not clear *a priori* which densities (orbital or total) should be used in the formulation[6,7]. Recently we have come to a resolution of this choice[8]. The various formulations of the OLPA were compared to the theoretical values obtained[9] for atomic targets from the dipole oscillator strength distributions of Inokuti *et al.*, and the formulation that best reproduces those numbers was chosen. The reliability of the results of Inokuti *et al.* has also been checked by calculating the mean excitation energies (and other moments of the dipole oscillator strength distribution) of He[10] and Be[11] using much more accurate methods, with indications that the Inokuti numbers may be a little (10%) too small. A further indication that the chosen procedure (Procedure 4 in the 1990 renewal proposal) works well comes from recent calculations on the Li hexagonal monolayer. As the lattice constant for this system goes from very large (20 au) to the equilibrium film distance, the mean excitation energy goes from the atomic (theoretical) to the solid (experimental) values[12]. Although not a completely definitive test of the method, either of the LPA or of its orbital decomposition, it is certainly a highly instructive one.

We have, so far, done only high accuracy benchmark calculations of stopping in molecules *via* polarization propagator calculations of the mean excitation energies of H_2[13], H_2O[13,14] and N_2[15]. These calculations can be used for later comparison of LPA-derived mean excitation energies. We have also calculated, in N_2, the anisotropic mean excitation energies, preparatory to a study of directionally dependent stopping in molecules and thin films.

A few years ago we realized that the electron velocity densities for target scatterers needed in the KT could be obtained from experimental Compton profiles[16] (CPKT). This allowed us to devise a semi-empirical scheme for extracting stopping powers from experimental data, and thus to avoid the difficult determinations of orbital (band) mean excitation energies and velocity densities normally encountered in our first principles work. The calculations carried out with this semi-empirical scheme, rather than being an integral part or basis of quality comparison for the first principles effort, have been used to point out and predict interesting trends in stopping

that might be addressed by experiment or, at some later stage, with the first principles theory. We mention a few of these applications below.

Using the scheme just outlined, we have studied the stopping of several solids based on experimental Compton profiles. We determined an effective experimental mean excitation energy for diamond, Si and Ge[17] and of some of the metals in the first transition series[18] by using the mean excitation energy as an adjustable parameter to match the CPKT stopping curve with the experimental stopping curve (in the region where the higher order Born corrections are not important). This scheme avoids the arbitrary choice of parameters such as shell corrections that normally plague the determination of mean excitation energies[19]. The result is a successful, potentially useful semi-empirical procedure. In the case of Li and Na[20], we carried out the same scheme, and compared the results to calculated atomic stopping cross sections[9]. Using the Ashley-Ritchie-Brandt form for the Barkas correction, we confirmed our earlier prediction[21] that there should be a large vapor/solid phase difference in the stopping in these systems, especially near the stopping maximum. We emphasize that the mean excitation energies determined in this way *are not used* in connection with the first principles calculations.

Using Compton profiles for localized chemical bonds and the CPKT scheme, we have calculated the Bethe stopping for a number of common bonds[22]. When added in the sense of the Bragg Rule to get molecular stopping, this scheme produces a better representation of the Bethe stopping of molecules than does the ordinary atomic Bragg Rule. Although not part of the first principles calculational effort, the scheme has been well received in the stopping community, and we are encouraged to attempt to put it on a first principles basis.

The primary goal of the research proposed in the 1990 renewal proposal is the calculation of stopping for various target systems, especially ultra-thin films. At the time the proposal was written, nearly a year ago, no published results were available. The preliminary results for the stopping of 1, 2 and 3 layer hydrogen square lattices in solid atomic and solid molecular configurations have now been completed. The work was reported first in invited talks at the Spring Meeting of the Danish Physical Society and later at the *11th Accelerator Conference*, and is now in press[5]. The results are very encouraging in that they reproduce physical and chemical trends as one moves from isolated atoms to systems more representative of condensed phase targets or to chemically bound systems. During the same period structural work has been completed on Li thin films up through 5 atomic layers thick[23]. Minimum energy geometries and properties have been determined, and the preliminary work on the stopping in the monolayer is under way[12]. Preliminary results are also encouraging in that they exhibit the predicted physical behavior, both for the mean excitation energy and stopping cross section.

All of the calculations reported to date are for normal incidence of the projectiles. As one of the areas of possible technological impact of this work is in space, where the incidence of particles is certainly not normal on any specific object, non-normal incidence must be considered. Recently we have considered non-normal projectile incidence on ultra-thin film targets, but the results are still too preliminary to report.

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